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KUM-104US PATENT #4 4-12-02

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Raman Naduhatty Selai	: Art Unit:
Serial No.:	(to be assigned)	: Examiner:
Filed:	(herewith)	:
FOR:	EXTRACTION OF AROMATICS FROM HYDROCARBON OIL USING FURFURAL-CO-SOLVENT EXTRACTION PROCESS	:

CLAIM TO RIGHT OF PRIORITY

Assistant Commissioner for Patents
Washington, D.C. 20231

S I R :

Pursuant to 35 U.S.C. 119, Applicant's claim to the benefit of filing of prior Indian Patent Application No. 25/Mum/2001, filed January 9, 2001, as stated in the inventor's Declaration, is hereby confirmed.

A certified copy of the above-referenced application is enclosed.

Respectfully submitted,

Allan Ratner, Reg. No. 19,717
Attorney for Applicant

AR/lk
Enc - Certified Copy of Indian Application

Dated: May 31, 2001

The Assistant Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

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Kathleen Libby

THE PATENTS ACT, 1970

IT IS HEREBY CERTIFIED THAT, the annex is a true copy of Application and Complete Specification filed on 9.1.2001 in respect of Patent Application No.25/Mum/2001 of Indian Oil Corporation Limited, a Public Limited Company, having its Head Office at G-9 Ali Yavar Jung Marg, Bandra (East) , Mumbai-400 051..

This certificate is issued under the powers vested on me under Section 147(1) of the Patents Act, 1970....

[redacted]

.....Dated this 27th day of April 2001.

(N.K.Garg)
Asst Controller of Patents & Designs

FORM 1

THE PATENTS ACT, 1970
(39 of 1970)
APPLICATION FOR GRANT OF A PATENT
(See sections 5(2), 7, 54 and 135 and rule 33A)

1. We, **INDIAN OIL CORPORATION LIMITED,** a Public Limited Company, having its Head Office at : G-9, All Yavar Jung Marg, Bandra (East), Mumbai 400 051.

hereby declare –

(a) that I am/we are in possession of an invention titled:

COVALENT FURFURAL EXTRACTION PROCESS FOR HYDROCARBON OIL

ORIGINAL

(b) that the **Complete specification** relating to this invention is filed with this application.

(c) that there is no lawful ground of objection to the grant of a patent to me/us.

2. Further declare that the inventor(s) for the said invention is / are :

1. Raman Naduhatty Selai
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

2. Devotta Irudayaraj
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

25/mum/2001
9/1/2001

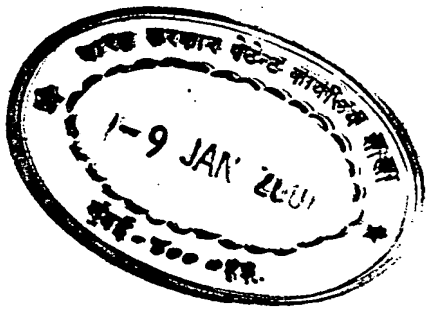
3. Bhaskar Mani
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

4. Venketesan Phoobalan
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

5. Rewat Bijendra Singh
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

6. Rawat Bachan Singh
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India

7. Bhatnagar Akhilesh Kumar
Indian Oil Corporation Limited
Research & Development Centre
Sector-13
Faridabad 121 007
Haryana
India



3. I/We, claim the priority from the application(s) filed in convention countries, particulars of which are as follows: NIL

4. I/We, state that the said invention is an improvement in or modification of the invention, the particulars of which are as follows and of which I/We are the applicant/patentee: NA

5. I/We, state that the application is divided out of my/our application, the particulars of which are given below: N.A.

6. That We are the assignee of the true and first inventors.

1. RAMAN NADUHATTY SELAI
2. DEVOTTA IRUDAYARAJ
3. BHASKAR MANI
4. VENKETESAN PHOOBALAN
5. REWAT BIJENDRA SINGH
6. RAWAT BACHAN SINGH
7. BHATNAGAR AKHILESH KUMAR

7. That our address for service in India is as follows:

KUMARAN & SAGAR
Trademark and Patent Attorneys
84C C-6 Lane (Off Central Avenue)
Sainik Farms
New Delhi-110 062, India
Telephone: 686 5955/6533 187/6533 182
Fax : 6533889, 6518717
E-mail: kns@ndb.vsnl.net.in

8. Following declaration was given by the inventor(s):

We the true and first inventors for this invention of or the applicant(s) in the convention country declare that the applicant(s) herein is/are my/our assignee or legal representative.

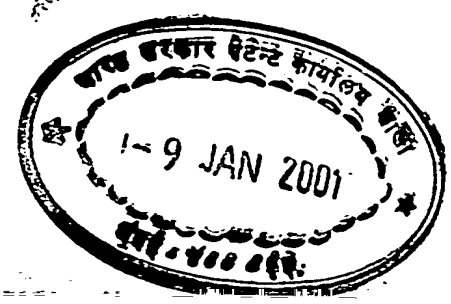
RAMAN NADUHATTY SELAI _____

DEVOTTA IRUDAYARAJ _____

BHASKAR MANI _____

VENKETESAN PHOOBALAN _____

REWAT BIJENDRA SINGH _____



RAWAT BACHAN SINGH

BHATNAGAR AKHILESH KUMAR

9. That to the best of my/our knowledge, information and belief the fact and matters stated herein are correct and that there is no lawful ground of objection to the grant of patent to me/us on this application.

10. Following are the attachment with the application:

Complete specification (3 copies)
Form 1 (3 copies)
Form 3 (2 copies)
Form 5 (2 copies)
Official fee of Rs.5000/-

I/We request that a patent may be granted to me/us for the said invention.

Dated this 6th day of January, 2001.

D. Gabriel

D.C.GABRIEL
OF KUMARAN & SAGAR
ATTORNEY FOR THE APPLICANT

To
The Controller of Patents
The Patent Office, Mumbai

FORM 2

THE PATENTS ACT, 1970

(39 of 1970)

ORIGINAL

COMPLETE SPECIFICATION

[See Section 10]

"COSOLVENT- FURFURAL EXTRACTION PROCESS FOR HYDROCARBON OIL"

INDIAN OIL CORPORATION LIMITED, a public limited company, G-9, Ali Yavar Jung Marg, Bandra (East), Mumbai 400 051.

The following specification particularly describes and ascertains the nature of this invention and the manner in which it is to be performed.

Field of the Invention

The invention relates to extraction of aromatics from hydrocarbon oils using cosolvent-furfural mixture to improve the selectivity. Use of this cosolvent-furfural mixture as solvent produces same quality raffinate as those of meat furfural measured by the refractive index, but produces higher yield of raffinate at same solvent to feed treat ratio.

Background and Prior Art references to the invention

Solvent extraction is a process that separates hydrocarbon mixtures into two phases, a raffinate phase which contains substances of relatively high hydrogen to carbon ratio often called paraffinic type materials and an extract phase which contains substances of relatively low hydrogen to carbon ratio often called aromatic type materials. Therefore, it may be said that solvent extraction is possible because different liquid compounds have different solution affinities for each other and some combinations are completely miscible while other combinations are almost immiscible. The ability to distinguish between high carbon to hydrogen aromatic type and how carbon is hydrogen or paraffinic type materials is termed selectivity. The more finely this distinguishing can be done the higher the selectivity of the solvent.

Solvent extraction of hydrocarbon oils using polar solvents to remove aromatic constituents has long been a standard processing procedure in the Oil Industry. The use of furfural to selectively extract aromatic components from a hydrocarbon oils is the subject of many patent, for instance, U.S. Patent No.2,079,885, U.S. Pat. No.2,698,276, U.S. Pat. No.3,567,627 and U.S. Pat. No.4,571,295, which are incorporated by reference. In U.S. Pat. No.5,922,193, ethers or aldehydes is added to furfural to improve the solvent capacity for debottleneeking the extraction unit and claimed 2-3vol% increase in the raffinate yield at solvent dosage of 250vol%. Further, in the said prior art, the recovery and material balance of the additive employed is not disclosed.

Objects of the invention

The main object of the invention is to provide an improved process for the production of lubricant base oil from an aromatic containing hydrocarbon oil by contacting the hydrocarbon oil with a solvent comprising furfural and a cosolvent.

Another object of the invention is to provide a method employing a cosolvent comprising aliphatic amide or mixtures of amides having an aliphatic carbon chain less than 5 carbon atoms.

Yet another object of the invention is to provide a method employing a cosolvent comprising aliphatic amide or mixtures of amides having an aliphatic carbon chain less than 3 carbon atoms.

Still another object of the invention is to provide a furfural extraction process for lube oil based stock production from hydrocarbon oils employing a cosolvent and furfural in a continuous countercurrent extraction column.

One more object of the invention is to provide a furfural extraction process with a cosolvent to facilitate phase separation and to increase raffinate yield, while maintaining the product quality as measured by raffinate refractive index.

Detailed description of the invention:

The present invention relates to an improved furfural extraction process for lube oil base-stock production from hydrocarbon oils by the addition of a cosolvent preferably, an aliphatic amide or mixture of amides. Addition of the cosolvent to furfural, conducted in a continuous countercurrent extraction column facilitates phase separation and increases raffinate yield while maintaining the same raffinate quality measured by raffinate refractive index. In this process, addition of a cosolvent, preferably an aliphatic amide or mixture of amides to furfural is done to facilitate phase separation and selectivity, wherein the raffinate yield increases by more than 3vol% and preferably, more than 5vol% at solvent dosage preferably less than 180vol% and more preferably less than 150% vol.

The invention therefore, includes an improved method for the production of lubricant base oil from an aromatic containing hydrocarbon oil comprising contacting the hydrocarbon oil with a solvent comprising furfural and a cosolvent, preferably an

aliphatic amide or mixture of amides having a aliphatic carbon chain less than five, and more preferably less than three under extraction conditions, producing a raffinate product with increased yield of more than 3vol% and preferably more than 5vol%. The invention further includes the cosolvent employed is recovered alongwith furfural and reused in the process.

In an embodiment, the improved furfural extraction process for hydrocarbon oils, which comprises contacting furfural extraction solvent in a unit with a cosolvent such as hereindescribed, to facilitate phase separation and to increase raffinate yield, while maintaining the product quality as measured by raffinate refractive index.

In another embodiment, the cosolvent is selected from an aliphatic amide or mixture of amides.

In yet another embodiment, the amides are selected from aliphatic series with carbon number less than five and preferably less than three.

In still another embodiment, the composition of amides in the solvent mixture is less than 30vol%, preferably less than 20vol% and more preferably less than 10vol%.

In another embodiment, the yield of raffinate increases by more than 3 vol% and preferably more than 5vol%.

In yet another embodiment, the solvent dosage is less than 250vol% preferably less than 180vol% and more preferably less than 150vol%.

In another embodiment, the cosolvent-furfural solvent mixture shows improved stability.

Feedstock

The process is applicable to hydrocarbon oils namely, vacuum gas oil, hydrotreated/hydrocaracked oil and catalytic cracker bottom boiling in the lubricant boiling range. The feedstocks may typically comprise hydrocarbons having initial boiling point of greater than 300°C and a final boiling point of about 600°C, preferably those fractions having a boiling range of about 370°C to 565°C. These distillate lubricant stocks namely, light neutral, interneutral and heavy neutral and are usually referred as solvent neutrals and are the distillate fractions of the vacuum tower.

Solvent extraction

Solvent extraction is conducted by contacting the lube distillate with a selective solvent, furfural in the present invention. Since a feedstock contains aromatics usually ranging from at least about 25 vol%, specifically from 25 to 80vol% and more specifically from 30vol% to 60vol%, the feedstock is initially subjected to an extraction step. Extraction utilizes a solvent, which is selective for aromatics, such as furfural in the present invention, and removes the aromatics, which contribute to poor stability and viscosity index. The solvent extraction is conducted with a solvent to oil ratio in the range of from about 0.5:1 to 10:1, such as in the range from about 0.75:1 to 5:1, depending on the feedstock. The operating conditions for furfural extraction cover a temperature range of about 25°C to about 175°C, preferably from about 50°C to 150°C. The yield in terms of volume percent typically ranges from 30 to 80. The characteristics of the product of solvent extraction are very important, and consideration of the solvent extraction conditions coupled with the choice of feed is necessary to achieve a product with the desired viscosity and VI, maximum yield of high VI product is achieved by adjusting the extraction severity. The resulting raffinate should have a VI of at least about 85, preferably 90. The aromatic-reduced raffinate should contain at most about 40vol% aromatics, preferably ranging from about 10 to 30vol%, even more preferably from 10 to 20vol%. The extractions may be preformed by conventional means, such as in a multistage countercurrent system, in a column with packing material or provided with perforated plates or in a column with a rotating shaft provided with dises.

Solvent

The process of the present invention involves the addition of a cosolvent preferably an aliphatic amide or mixture of amides to furfural to facilitate phase separation and selectivity. The process of the present invention involves the addition of small volumes of one or more cosolvents to furfural to increase raffinate yield. The properties of the cosolvent employed for the present invention are listed in Table 1.

Table 1
Cosolvent Properties

S.No.	Name of the solvent	Melting Point °C	Boiling Point °C	RI	Density @20°C gm/ml
1	Formamide	2-3	210	1.447	1.134
2	N- Methyl Formamide	-4	198-199	1.432	1.001
3	N,N Di Methyl Formamide	-61	153	1.431	0.945
4	Acetamide	79-81	221	-	-
5	N-Methyl Acetamide	26-28	204-206	1.433	0.957
6	N,N- Di Methyl Acetamide	-20	164.5-166	1.438	0.937
7	Propionamide	80-83	213	-	1.042
8	N-Methyl Propionamide	-43	79	1.377	0.915
9	N,N Di Methyl Propionamide	-45	174-175	1.440	0.920

Generally, the co-solvent is added in an amount less than about 30vol% based on total solvent such as less than about 20vol% based on total solvent, less than about 10vol% based on total solvent and less than about 5vol% based on total solvent, depending on the feedstock. For example, a 10vol% co-solvent 90vol% furfural blend may be used in the extraction process of the present invention when the feedstock is Arab mix Interneutral distillate. Cosolvents used in the process of the present invention also have a boiling point in the range of from about 50°C to 225°C, preferably in the range of from about 75°C to 200°C. The addition of co-solvents, such as N-Methyl Acetamide to furfural improves its selectivity for extraction of aromatics from lube distillates. Use of cosolvents infurfural extraction increases the raffinate yield at the same raffinate refractive index (RI).

In essence, the invention includes an improved method for the production of lubricant base oil from an aromatic containing hydrocarbon oil comprising contacting the hydrocarbon oil, with a solvent comprising furfural and a cosolvent, preferably an aliphatic amide or mixture of amides having a aliphatic carbon chain less than five and more preferably less than three under extraction conditions, producing a raffinate product with increased yield of more than 3vol% and preferably more than 5vol%. The invention further includes the cosolvent employed is recovered along with furfural and reused in the process.

The advantage of the present invention allows for retrofitting existing equipment. An additional advantage of the furfural/co-solvent mixture of the present invention is the lower operating cost as the cost of the co-solvent employed is lower than that of neat furfural. The addition of the co-solvent of the present invention also improves the stability of the resultant co-solvent-furfural blend compared to furfural alone preventing degradation of furfural, which results in lower furfural loss.

Description of the preferred embodiment

The following examples illustrate the process of the present invention. Arab mix Interneutral distillate having the properties as set forth below in Table 2, was used for each extraction example.

Table 2
Properties of Inter-Neutral Distillate

S.No.	Properties	Result
1	Density @ 15°C, gm/ml	0.9385
2	API Gravity	19.3
3	Refractive Index at 60°C	1.50197
4	Kinematic Viscosity, cSt at 60°C	50.07
	100°C	9.25
5	Distillation (ASTM D 1160), °C	
	IBP/ 5 %vol rec.,	364/405
	50/90	471/519
	95	537

For each furfural, with or without co-solvent, continuous countercurrent extraction was performed in a three meter height bench scale jacketed glass column extraction apparatus. The feed, Arab mix distillate was heated and pumped for example. 1.0kg hr from bottom of the extractor (feed inlet) and the solvent, furfural with or without cosolvent were heated and pumped for example, 1.5kg/hr at the top of the extractor (solvent inlet). The solvent rate was varied accordingly to the desired solvent to freed weight ratios of 1:1.5 and 1:1.8. (These ratios are typically referred to as 150% and 180% solvent dosage). The extractions were performed at column top temperature for example, 110°C and column bottom temperature for example 70°C. After the mixture of solvent and oil pumping started from their respective inlets, the raffinate phase from the top of the column

(raffinate product phase outlet) and extract phase from the bottom of the column (extract product phase outlet) were drawn continuously. The interface between the lighter raffinate phase and heavier extract phase flow. After steady state, say, after 1 to 2 hrs, indicated by constant interface level and constant raffinate and extract phases flow, the raffinate phase and the extract phase were collected simultaneously for a given period of time say, 30 to 40 minutes, in all three to four batches. The two phases collected were weighed to ensure material balance closure. The solvent was stripped from the extract and raffinate with nitrogen under vacuum. The stripped raffinate and extract phases were weighed and the raffinate yield was obtained. Final raffinate samples were analyzed for, Density, API gravity, and Refractive Index (RI).

The results from the continuous countercurrent extraction are shown below as Example 1 and Example 2 corresponding to two Solvent to Feed ratio's (S/F ratio) 1.5 and 1.8, respectively while the rest of the operating parameters are kept constant.

Commercially, lube extraction units are operated to a RI specification since for a particular lube crude and type of refining process, raffinate RI correlates with the viscosity index (VI) of the dewaxed oil (DWO), with lower RI corresponding to higher VI. Analysis of the data in Example 1 shows that for extraction conducted at solvent to feed treat ratio of 1.5 or solvent dosage of 150vol%, the cosolvent-furfural blends are more effective than furfural alone, resulting in more than 3 vol% improvement in raffinate yield at same raffinate RI. Similarly, the Example 2 shows an increase of more than 5vol% of raffinate yield at same RI at solvent to feed treat ratio of 1.8 or solvent dosage of 180vol%.

Example 1
Countercurrent extraction experimental conditions and Raffinate Properties

Properties	Furfural	Furfural with Cosolvent
Experimental conditions		
Column Top temperature, °C	110	110
Column Bottom temperature, °C	70	70
S/F ratio (wt/wt)	1.5	1.5
Raffinate Properties		
Raffinate Yield, vol%	67.8	71.2
RI at 60°C	1.4729	1.4727
Density @15°C, gm/ml	0.8874	0.8871
API Gravity	28.0	28.0
Extract Properties		
Density @15°C, gm/ml	1.0300	1.0305
API Gravity	5.9	5.8

Example 2
Countercurrent extraction experimental conditions and Raffinate Properties

Properties	Furfural	Furfural with Cosolvent
Experimental conditions		
Column Top temperature, °C	110	110
Column Bottom temperature, °C	70	70
S/F ratio (wt/wt)	1.8	1.8
Raffinate Properties		
Raffinate Yield, vol%	63.8	69.7
RI at 60°C	1.4701	1.4697
Density @15°C, gm/ml	0.8835	0.8827
API Gravity	28.7	28.8
Extract properties		
Density @15°C, gm/ml	1.0243	1.0248
API Gravity	6.6	6.6

WE CLAIM

1. An improved furfural extraction process for hydrocarbon oils, which comprises contacting furfural extraction solvent in a unit with a cosolvent such as hereindescribed, to facilitate phase separation and to increase raffinate yield, while maintaining the product quality as measured by raffinate refractive index.
2. A process as claimed in claim 1 wherein the cosolvent is selected from an aliphatic amide or mixture of amides.
3. A process as claimed in claim 2 wherein, the amides are selected from aliphatic series with carbon number less than five and preferably less than three.
4. A process as claimed in claim 2 wherein, the composition of amides in the solvent mixture is less than 30vol%, preferably less than 20vol% and more preferably less than 10vol%.
5. A process as claimed in claim 1 wherein the yield of raffinate increases by more than 3 vol% and preferably more than 5vol%.
6. A process as claimed in claim 1, wherein the solvent dosage is less than 250vol% preferably less than 180vol% and more preferably less than 150vol%.
7. A process as claimed in claim 1, wherein the cosolvent-furfural solvent mixture shows improved stability.
8. An improved furfural extraction process for hydrocarbon oils substantially as hereindescribed and illustrated with reference to the examples.

Dated this 6th day of January, 2001.

D. Caleb Gabriel

D.C.GABRIEL
OF KUMARAN & SAGAR
ATTORNEY FOR THE APPLICANT

ABSTRACT

An improved furfural extraction process for lube oil base-stock production from hydrocarbon oils by the addition of a cosolvent and conducted in a continuous countercurrent extraction column that facilitates phase separation and increases raffinate yield while maintaining the same raffinate quality measured by raffinate refractive index.